



## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Kazuo ISHIWARI et al. Conf.: 9616  
Appl. No.: 09/787,303 Group: 1772  
Filed: March 16, 2001 Examiner: J. Rhee  
Title: POLYTETRAFLUOROETHYLENE BLOCK-SHAPED MOLDED  
ARTICLE AND METHOD OF PRODUCING THE SAME

DECLARATION UNDER 37 C.F.R. § 1.132

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I, Masahiko YAMADA, a citizen of Japan, residing at c/o Yodogawa Works of Daikin Industries Ltd., 1-1, Nishihitotsuya, Settsu-shi, Osaka-fu, Japan declare and say as follows:

1. I am one of the co-inventors of the above-identified application.
2. I graduated from Nihon University, College of Science and Technology, Department of Mechanical Engineering in March 1988.
3. Since December 1996 up to the present time, I have been employed by Daikin Industries, Ltd. and engaged in research works on the development of a molding process of a fluororesin.
4. I had performed the following experiments and beg to submit herewith the exact report thereon.

**(A) Conventional procedure****EXPERIMENT A-1** (Melt viscosity:  $6.55 \times 10^9$  poise)

(the same as Comparative Example 1 of the present Description)

A powder (average particle size: about 30  $\mu\text{m}$ ) of tetrafluoroethylene /perfluoropropyl vinyl ether copolymer (melt viscosity at 380°C of the copolymer:  $6.55 \times 10^9$  poise) obtained by suspension polymerization was compression-molded at 25°C under a pressure of 200 kg/cm<sup>2</sup> for 120 minutes to give a preform as shown in Fig. 1 of the present application. The preform had a length L of about 100 cm, and a diameter D of about 42 cm. The diameter of a hole was about 15 cm.

The preform was baked by means of a conventional method (longitudinal baking), in which the preform was heated while the preform was positioned in a furnace so that the rotation symmetry axis of the preform was in the gravitation direction. The preform was kept at a temperature of 340 to 380°C for 50 hours. The load per unit area at the time of baking the preform ((weight of the preform (250 kg))  $\div$  (bottom surface area of the preform 1,210 cm<sup>2</sup>)) was 207 g/cm<sup>2</sup>. The baking gave a block-shaped molded article. The block-shaped molded article had a length of about 100 cm, a diameter of about 42 cm, and a diameter of the hole of about 15 cm.

The weight loss (block deformation amount) until a stable film or sheet could be cut from the block-shaped molded article was 7.0%. This datum is plotted in Fig. A which shows relationship between the block length (that is, block height) and the deformation amount.

A photograph of the resultant block-shaped molded article is shown in Fig. B (left side).

**EXPERIMENT A-2** (Melt viscosity:  $6.55 \times 10^9$  poise)

The same procedure as in Experiment A-1 was repeated except that the size of the preform was changed. The results are shown in Fig. A.

**EXPERIMENT A-3** (Melt viscosity:  $1.32 \times 10^{10}$  poise)

The same procedure as in Example A-1 was repeated except that the tetrafluoroethylene/perfluoropropyl vinyl ether copolymer had a melt viscosity at 380°C of  $1.32 \times 10^{10}$  poise and the size of the preform was changed. The results are shown in Fig. A.

**(B) The present invention procedure**

**EXPERIMENT B-1** (Melt viscosity:  $6.55 \times 10^9$  poise)

(the same as Example 2 of the present Description)

A powder (average particle size: about 30  $\mu\text{m}$ ) of tetrafluoroethylene /perfluoropropyl vinyl ether copolymer (melt viscosity at 380°C of the copolymer:  $6.55 \times 10^9$  poise) obtained by suspension polymerization was compression-molded at 25°C under a pressure of 200 kg/cm<sup>2</sup> for 120 minutes to give a preform as shown in Fig. 1 of the present application. The preform had a length L of about 100 cm, and a diameter D of about 42 cm. The diameter of a hole was about 15 cm.

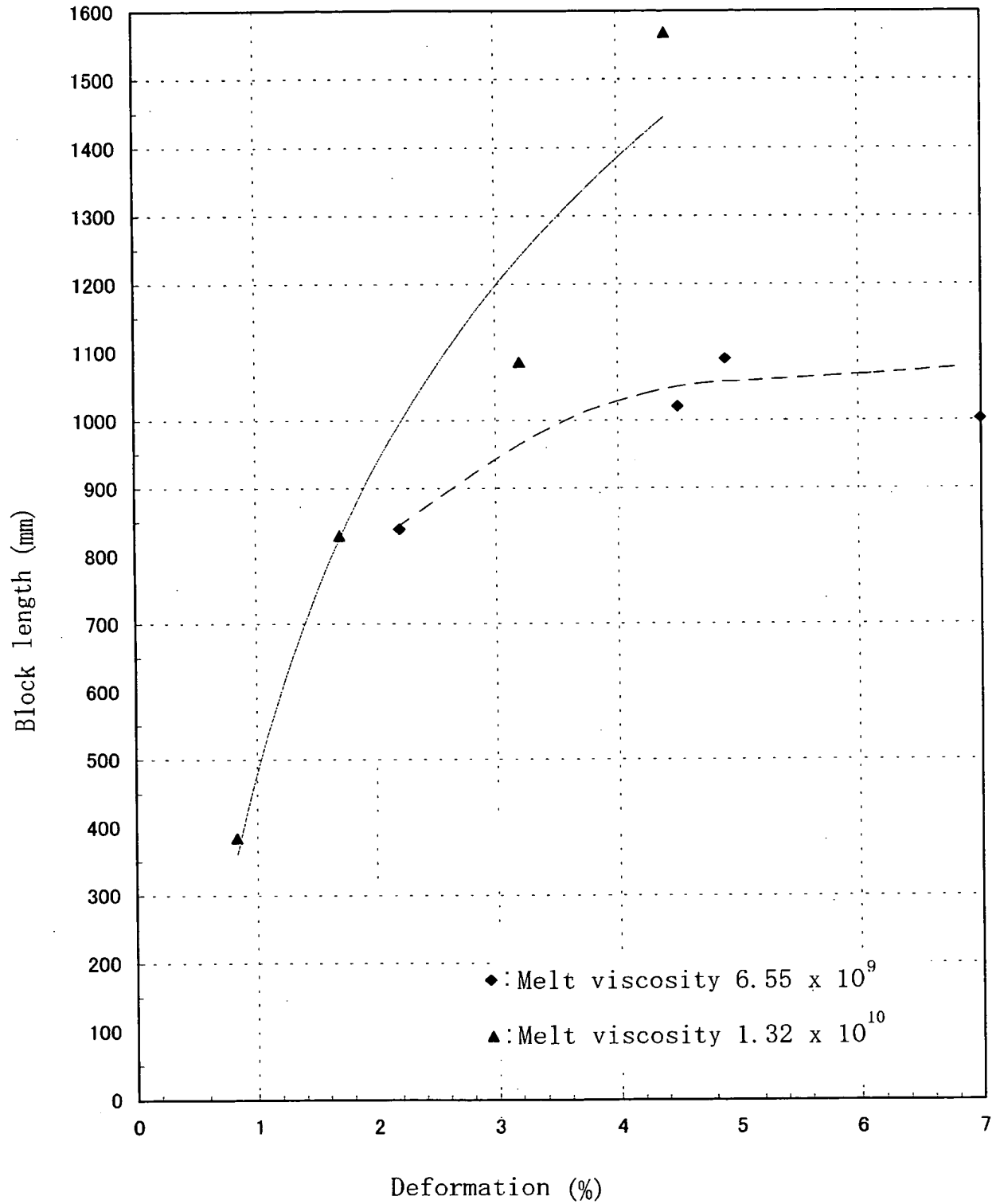
The preform was baked in the manner shown in Fig. 2 of the present application. The outer diameter of the cross-section of the two rolls was 15 cm. With respect to the stainless steel pipe outside of the molded article, the outer diameter was 50 cm, and the thickness was 1 cm. With respect to the stainless steel pipe inside of the molded article, the outer diameter was 12 cm, and the thickness was 1 cm. The rotation speed of the rolls was adjusted so that the rotation speed of the preform was 90 revolutions per hour. The baking was carried out, while rotating the preform at 90 revolutions per hour and keeping the preform at a temperature of 340 to 380°C for 50 hours. The load per unit area at the time of baking the preform [(weight of the preform (250 kg))  $\div$  (outer surface area of the preform excluding the two bottom surface areas (i.e. the area of the surface of the preform brought into contact with the pipe at the baking time) (13,190 cm<sup>2</sup>))] was 19 g/cm<sup>2</sup>. The baking gave a block-shaped molded article. The block-shaped molded article had a length of about 108 cm, a diameter of about 40 cm, and a diameter of the hole of about 14 cm.

The weight loss (block deformation amount) until a stable film or sheet could be cut from the block-shaped molded article was 0.6%.

A photograph of the block-shaped molded article is shown in Fig. B (right side).

**Fig. A**

Block length vs Deformaion amount  
Conventional baking



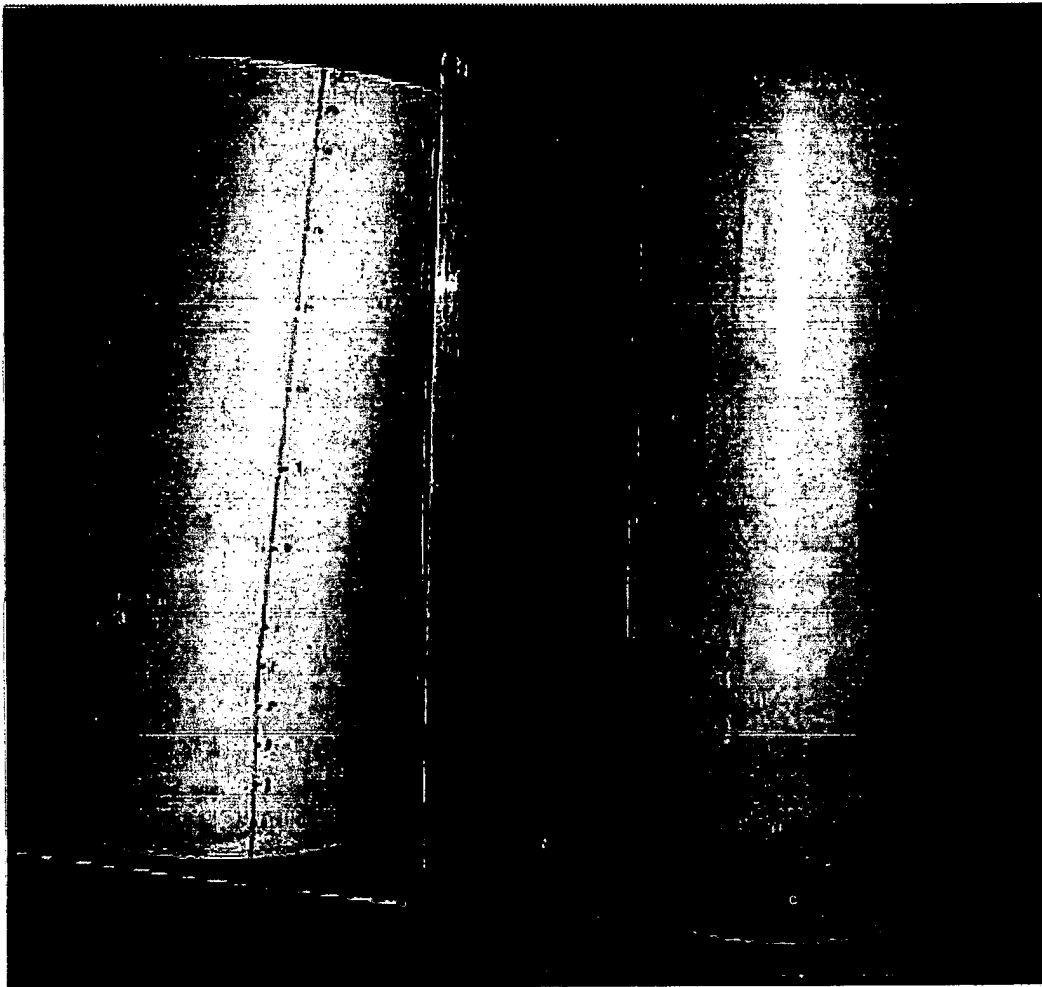


**Fig. B**

**Photographs of the block-shaped molded articles**

Experiment A-1  
(Conventional)

Experiment B-1  
(Present invention)



## Discussion

### (1) Results of Fig. A

Fig. A shows the relationship between the block length (that is, block height) and the deformation amount of the molded article prepared by conventional procedure.

When the articles having the same melt viscosity of  $1.32 \times 10^{10}$  poise are compared, it is indicated that the larger the article is, the larger the deformation amount (%) is. Stated differently, the deformation amount (%) of the larger article is much larger than the deformation amount of the smaller article, when the articles have the same melt viscosity.

When the articles having the same block length are compared, it is indicated that the larger the melt viscosity is, the smaller the deformation amount (%) is. Stated differently, the deformation amount (%) of the article having larger melt viscosity is smaller than the deformation amount of the article having smaller melt viscosity, when the articles have the same size.

### (2) Results of Fig. B

Fig. B shows a photograph of the molded articles prepared by the conventional procedure (left side) and the present invention procedure (right side). It is clear that the conventional procedure article has much larger deformation than the present invention procedure article.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S. Code 1001 and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Masahiko Yamada  
Masahiko YAMADA

Dated this 19<sup>th</sup> day of November 2003